Investigation of Microwave-Induced Chemical Etching in CR-39

Jayden Roberts

Brockport High School

Brockport, NY

Advisor: Steven Ivancic

Laboratory for Laser Energetics

University of Rochester

Rochester, NY

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Abstract

CR-39 is a solid-state nuclear track detector that can be used to collect information on critical performance metrics of inertial confinement fusion implosions, such as the areal density of the confined fuel. Charged particles emitted from reactions occurring inside the target damage polymer bonds in the CR-39. These damages are on the scale of nanometers, and thus require further treatment to be recorded by instruments such as optical microscopes. LLE currently uses a several-hour chemical etching process to develop particle tracks. Microwave radiation has been proposed to work in tandem with chemical etching to greatly increase the rate at which tracks develop. A study of microwave-induced chemical etching was undertaken and found to fully develop alpha particle tracks in CR-39 in 30 minutes as opposed to 6 hours in conventional chemical etching used today.

Introduction

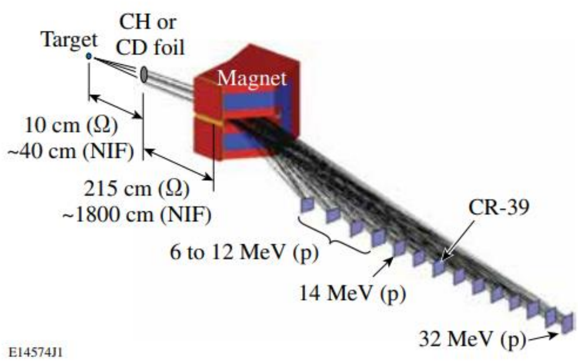


Figure 1: A diagram of the Magnetic Recoil Spectrometer. Energetic neutrons resulting from the fusion implosion collide with a CH or CD foil, producing energetic protons or deuterons. These charged particles pass through a magnet, causing them to be deflected by the Lorentz force onto a series of CR-39 pieces. The deflection angle is inversely proportional to the particle's velocity, allowing the particle's energy to be deduced by its position. Figure from Ref. 2.

Nuclear fusion is the process of fusing two lighter nuclei into a heavier nucleus, releasing energetic particles in the process. Targets can be manufactured with a variety of isotopes, but deuterium and tritium are the preferred pair at the Laboratory for Laser Energetics (LLE) for their energy efficiency. For relevant information to be gleaned from a fusion implosion, diagnostic instruments must be fielded to capture data from the target. The Magnetic Recoil Spectrometer (MRS)[1,2]—shown in Figure 1—is one of many such diagnostics fielded on the OMEGA laser at LLE. The MRS is used in conjunction with other charged-particle diagnostics to infer parameters such as areal density, which is calculated by the equation:

$$\rho R = \int_0^R \rho(r) dr,$$

where $\rho(r)$ is the density at radius r. The areal density quantifies the quality of the compression of a target during an implosion.

Energetic neutrons from the fusion implosion collide with a CD or CH foil on the front of the MRS diagnostic, releasing a charged particle known as a recoil deuteron or recoil proton, respectively. This charged particle then travels through a magnetic field, which deflects the particle onto one of a series of CR-39 pieces. The piece absorbs the energetic particle, leaving nanoscale damage—referred to as latent tracks. These pieces are then recovered from the diagnostic and must undergo further development to enlarge the track pits to the microscale. The currently preferred method for this development is chemical etching.

Chemical etching works by introducing an etchant, usually a basic solution such as sodium hydroxide, to the CR-39 piece at a high temperature. This high temperature allows the etchant to attack the polymer bonds damaged by the charged particles. This mechanism has been explored in the literature and is the result of the etchant preferentially attacking regions with free radicals displaced by the charged particles[3]. This process, given sufficient time, results in the conical pits shown in Figure 2.

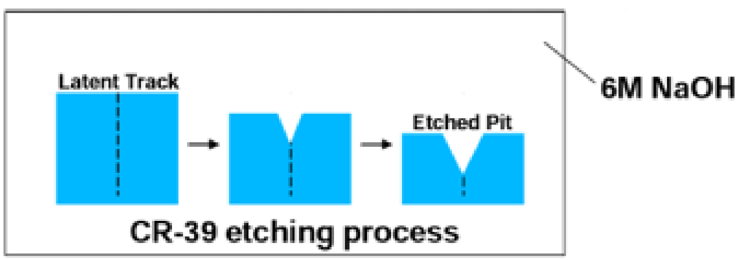


Figure 2: Etching of CR-39. Charged particles are slowed down inside of the CR-39, leaving behind a trail of broken polymer bonds—referred to as latent tracks. These tracks can then be developed using an etching process, revealing conical pits.

It has been shown in the literature that the presence of microwaves can speed up the etching process. This new method is called microwave-induced chemical etching (MICE)[3,4] and has been shown to substantially increase the rate of etching.[4] Though referenced literature has introduced MICE as a means of developing latent tracks on CR-39, further investigation is required to implement such a method at LLE.

Methods and Materials

The experimental setup is shown in Figure 3. The setup consists of a variable-power 1000-Watt microwave, four 400 mL Teflon beakers, and a thermometer.

Figure 3: The experimental setup. Teflon beakers and lids are used to ensure a microwave-safe vessel for the etchant. The thermometer is placed in a beaker of water after temperature measurements are taken to prevent a buildup of etchant on laboratory surfaces.

The etchant, 6 N sodium hydroxide, was decanted into four beakers, with each beaker containing 200 mL of etchant. The beakers were then covered with lids to minimize evaporation, though a small opening was kept, ensuring that there would be no build-up of pressure resulting from vapor. These beakers were then placed into a variable-power microwave and microwaved for two minutes. The beakers were then removed from the microwave and the etchant was stirred to ensure a uniform temperature distribution. The temperatures of all four solutions were measured, and the beakers were placed back into the microwave for another two minutes. This procedure was carried out until the solution reached 100°C. This process has been standardized in the literature[3]. The results of this procedure can be seen in Figure 4, which plots temperature against time for varying power settings. The plotted temperature readings were calculated by taking the average temperature of all four beakers.

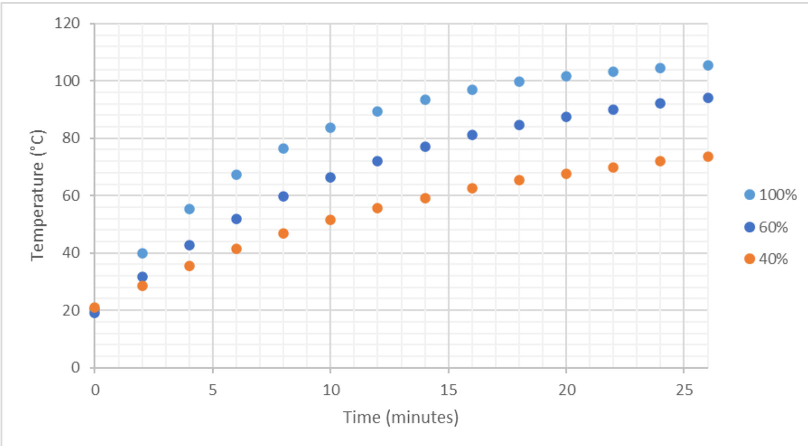


Figure 4: A summary of temperature over time for varying power settings. The beakers initially warm up quickly but temperatures eventually plateau due to the equilibrium between heating and cooling. The former occurs because the solutions are being microwaved, and the latter results primarily from the beakers being removed from the microwave for a temperature reading.

To use this data for etching, we must first account for the cooling during temperature measurements. This cooling would not occur if the beakers were simply left in the microwave. We fit a differential equation modeling constant heating and Newtonian cooling to resolve this issue:

$$\frac{dT}{dt} = s - k(T - a),$$

where s represents the constant rate of increase of temperature due to the microwave heating source, k represents the resulting cooling, T is the modeled temperature of the beakers that is being solved for, t is time, and a is the room's ambient temperature. This differential equation can be solved by separation of variables, giving the result:

$$T(t) = a + c_1 e^{-kt} + \frac{s}{k},$$

where c_1 is dependent on initial conditions. Substituting $t = 0$ yields

$$T(0) = a + c_1 + \frac{s}{k},$$

which we know is the ambient temperature of the room; thus,

$$c_1 = -\frac{s}{k}, \text{ giving,}$$

$$T(t) = a + \frac{s}{k}(1 - e^{-kt}),$$

which can then be used to fit our data. To do this, a Python program was written using the curve fit function in SciPy[5]. The resulting parameters are given in Table 1 and the fitted curve is shown in Figure 5.

Power Setting	$\boldsymbol{s}\ (\frac{^\circ C}{s})$	$\boldsymbol{k}\ (\frac{1}{s})$
40%	4.4	0.07
60%	6.7	0.08
100%	11	0.1

Table 1: Heating and cooling parameters, s and k respectively, for various power settings. As expected, the fitted values of s are proportional to the power setting of the microwave. It was expected that k would be approximately constant regardless of power setting, which is the case. The variation seen is due to measurements taking slightly longer by random chance.

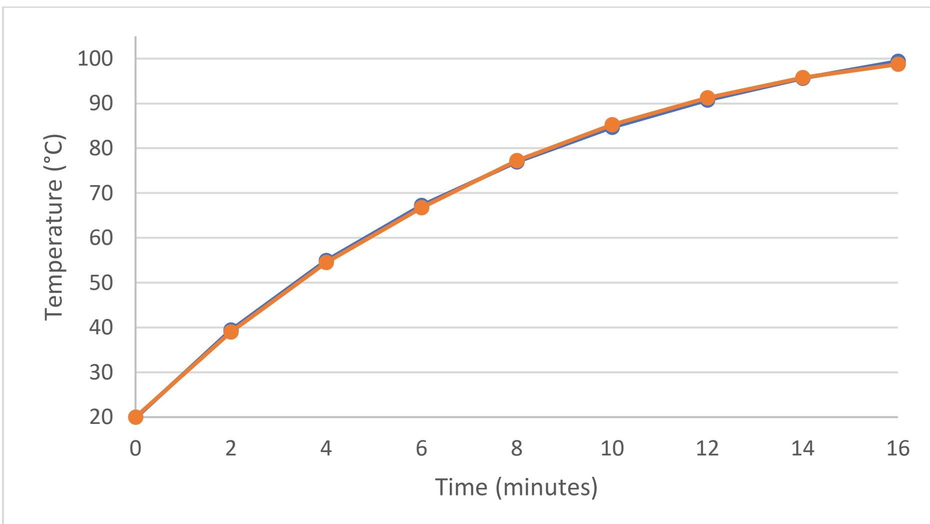


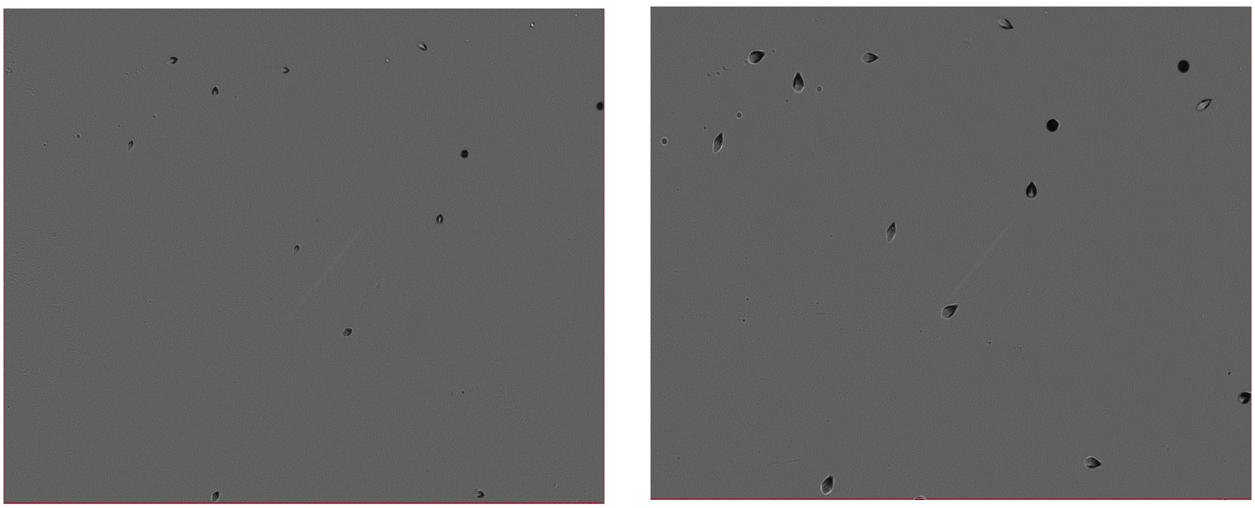
Figure 5: The recorded and modeled temperatures plotted over time for the 100% power setting. The orange data plots are the recorded data and the blue data points are the modeled temperatures calculated using the parameters in Table 1.

For optimal etching conditions, the etchant must be first heated to 70°C. Then, a step of etching can be carried out until the solution reaches 100°C. Etching must stop at this temperature to ensure data integrity[4]. The etchant is then allowed to cool back to 70°C. This was done by taking intermittent temperature measurements in conjunction with a Newtonian cooling model to approximate how long cooling will take. This step etching process can be repeated until the etch duration has elapsed. This corresponds to step etching durations of 7 minutes, 4 minutes, and 3 minutes for 40%, 60%, and 100% power levels respectively, with a 4-minute preheating period at 100% power to reach the necessary 70°C for etching. These durations were calculated directly from the constant rate of increase of temperature from the microwave.

The center of a piece of CR-39 was irradiated with an Americium alpha particle source for five seconds at a distance of 0.5 cm. The alpha particles did not pass through a collimator. The beakers of etchant were then brought to 70°C per the previously outlined process. The piece of CR-39 was then placed into a beaker and microwaved for the prescribed 3-minute interval. After this time the piece was scanned, and the average particle pit diameter was measured.

Results

A region of the CR-39 was captured by an optical microscope at various times, showing the growth of alpha particle pits. Typical images are shown in Figure 6. Further inspection via the microscope revealed a uniform development of alpha tracks throughout the central region of the piece.



a. b.

Figure 6: Developing alpha particle tracks on CR-39. (a) A piece of CR-39 irradiated with alpha particles after 12 minutes of etching. (b) The same piece after 30 minutes of etching, where the pits are fully developed. The pieces were examined under the same optical microscope in the same region to ensure consistency.

Furthermore, the oblique incidence of the alpha particles is revealed upon inspection. This geometry corresponds to our expectation of tracks from an uncollimated alpha source. This is an important first step in ensuring the signal integrity of a piece prepared using MICE. Following the MICE procedure, we develop CR-39 pieces in 30 minutes, as opposed to the 6 hours required for chemical etching.[6]

Conclusion

This project investigated the merit of using microwave-induced chemical etching as an alternative to traditional chemical etching methods. The goals were to outline the process of microwave-induced chemical etching and supply a comparison between the duration necessary for the development of alpha particle pits. MICE was found to be a superior option, with traditional methods taking 6 hours to achieve fully developed alpha particle tracks whereas MICE develops latent particle tracks after 30 minutes.

The mechanism by which microwaves influence etching has been proposed in the literature[3]. Microwaves heat objects when dipoles within the object interact with microwave radiation, giving rise to dielectric heating. This mechanism favors polar regions, such as those with free radicals created by the alpha particles. This enables the etchant to more favorably attack latent particle tracks, giving rise to a shorter etch duration.

Further areas for exploration with MICE include testing for uniform development of pits throughout all regions of the CR-39 piece, and using recoil protons instead of alpha particles—which would entail an experiment involving a CH or CD foil along with a neutron source. Additionally, a comparison of the signals on pieces prepared using traditional methods and MICE would be integral. These experiments would serve to prove MICE as a reliable alternative in more than just duration.

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